PERFORMANCE AND DURABILITY OF HgI_2 X-RAY DETECTORS FOR SPACE MISSIONS

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Abstract

Considerable recent progress has been achieved in HgI_2 detector fabrication technology and amplification electronics. An energy resolution of 198 eV (FWHM) has been obtained for the Mn k_α line of 5.9 keV in a practical x-ray probe without the use of cryogenic cooling. Detectors prepared with Parylene-C encapsulation have demonstrated perfect reliability in two-year tests under high vacuum, and temperature and bias cycling. Other HgI_2 detectors were used to demonstrate proton radiation damage resistance to levels of 10^{12} protons/cm^2 at 10.7 MeV.

Introduction

A key element in x-ray spectroscopy systems, particularly for arrays of light elements, is the quality of the energy dispersive detector. A system energy resolution of 140 to 200 eV (FWHM) at 5.9 keV is typically required depending on the application. To date, this ordinarily has required the use of cryogenically cooled silicon or germanium x-ray detectors. Recent work has shown that the energy resolution for a mercuric iodide (HgI_2) spectrometer can approach that of silicon or germanium spectrometers.

A major advantage to the use of a HgI_2 system in many applications is that it does not require liquid nitrogen for cooling (1-3). It is this advantage that has led to a program to evaluate and develop such a system for the Scanning Electron Microscope and Particle Analyzer (SEMPA) instrument (4,5,8) being developed for NASA's Mariner Mark II Comet Rendezvous/Asteroid Flyby Mission (6). This mission has a planned duration in excess of seven years and thus makes considerable demands on long term reliability of the spacecraft as well as science instruments such as SEMPA. At the start of the program the ultimate ability of HgI_2 detectors to meet both the SEMPA resolution requirement (200 eV) and the longevity requirement was quite uncertain.

Significant progress has been achieved in HgI_2 detector performance through improvements both in fabrication technology and low noise amplification electronics. Significantly improved spectral response has been achieved with the introduction of a guard-ring detector contact and a collimating metal shield close in front of the detector. Especially at high count rates, these two advancements have minimized noise contributions from charge generation and collection in regions with weak electric fields (2,7). Reduction in preamplifier electronic noise has been achieved through utilization of newly developed FET structures made by the Interfet Corporation. One new FET, type SNJ14L03, has a geometry with a better figure of merit (i.e., the ratio of transconductance to input capacitance, g_m/C_i) for low capacitance detectors and thus gives lower noise for our application.

Spectral Resolution

The SEMPA laboratory research prototype, shown in Figure 1, has been a test-bed for the evaluation of improvements to practical detector systems. The x-ray probe system used for ultimate resolution tests is in the lower left part of Figure 1. The details of the probe system are shown schematically in Figure 2. The HgI_2 detector used had an active area of 5 mm^2.

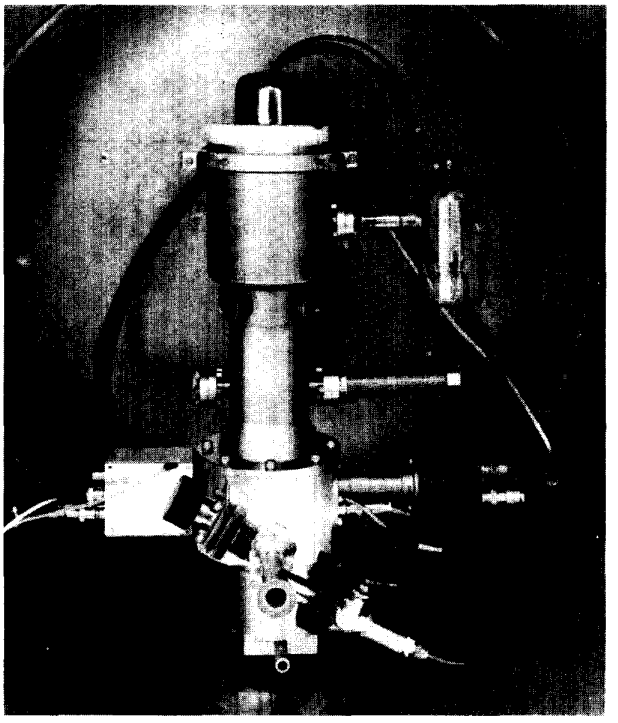

Figure 1. The SEMPA (Scanning Electron Microscope and Particle Analyzer) laboratory research prototype.

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It was mounted 10 mm from the target and intercepted 0.05 steradian solid angle. The detector leakage current was less than 0.1 pA and the detector capacitance was less than 1 pF. The system uses a single stage thermoelectric cooler to reduce the detector temperature to about $0^{o}C$. The preamplifier input FET is cooled to about $-40^{o}C$ with a three stage thermoelectric cooler. The detector is protected from backscattered 15 keV electrons by an 8 μm Be window. The detector system is exposed to the vacuum environment of the SEM column. The typical system operating pressure of 3×10^{-7} Torr is achieved by a liquid nitrogen trapped diffusion pump. In this thermoelectrically cooled HgI_2 spectrometer system a

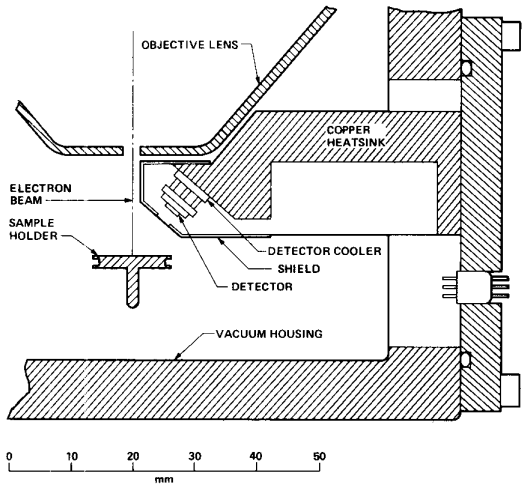

Figure 2. General configuration of the HgI_2 detector in the SEMPA instrument.

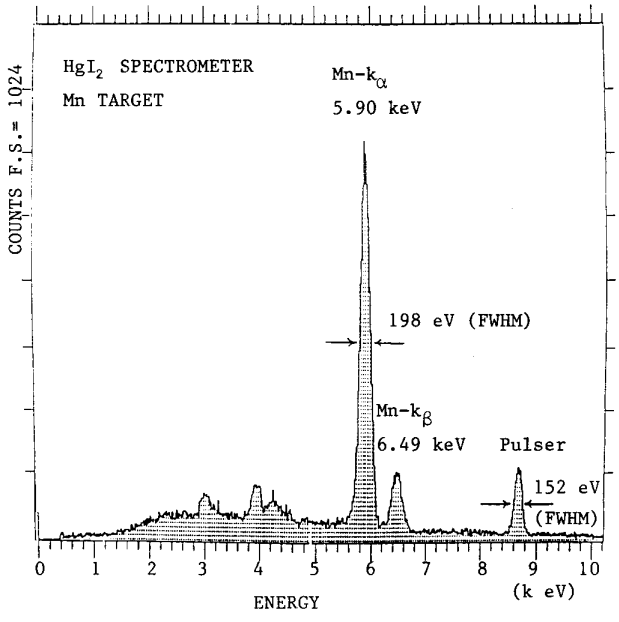

Figure 3. Manganese spectrum taken with the HgI_2 SEMPA prototype detector.

best-ever, total resolution of 198 eV (FWHM) was obtained for the 5.9 keV k_α line of Mn, Figure 3. The electronic noise contribution for this system was about 152 eV (FWHM) measured by pulser method. The main amplifier was a Tennelec TC244. The triangular shaping mode was chosen with the peaking time of 28.8 μs.

Detector Encapsulants

Proper HgI_2 surface passivation and device encapsulation are critical for insuring long term detector reliability. Although unprotected mercuric iodide crystals do not exhibit gross short or long term effects when exposed to normal laboratory storage environments, various gases, vapors, and particularly a vacuum environment, can rapidly, adversely affect detector performance. Several excellent protective surface treatments have been identified to date. The materials which have undergone initial development and testing are silicones, acrylics such as Conap CF-1170 and polymethylmethacrylate (PMMA), and Parylene (a Union Carbide product).

Some room temperature curing silicone compounds that worked well in a laboratory atmosphere provided little protection against HgI_2 evaporation in the vacuum, perhaps because the vacuum removed moisture from the compounds. Some coatings, especially acrylics, which are applied in a solution and harden with solvent evaporation, have proven to be chemically compatible with HgI_2 and an excellent barrier to HgI_2 evaporation. However, applying an appropriately thin coating to a detector from any solvent based system was found to be impractical for two major reasons: First, most solvent based systems have the problem that HgI_2 is significantly soluble in the solvent. Second, it was difficult to control the thickness of the coating so that, simultaneously, the coating over the active area was thin, while the coating at the edges was thick enough to prevent evaporation. The solubility of the HgI_2 resulted in the applied coatings always containing small amounts of HgI_2 that produce noticeable x-ray absorption. Typically, the difficulty in thickness control resulted in detectors that had significantly reduced low energy sensitivity due to excess coating thicknesses on the active areas, but were still poorly coated at the edges (3).

Parylene coatings have several desirable attributes, including their method of application, polymerization and deposition from a vapor, which allows for a very uniform and well controlled coating thickness even in submicron layers. Corners and edges are typically coated to the same thickness as are open surface areas. The low atomic number of some Parylenes make them useful as x-ray transparent windows. The most extensively tested has been polymerized dichloro-di-1,4 xylylene (Union Carbide Parylene-C). Tests of coating performance have included storage of coated HgI_2 crystals at elevated temperatures ($75^{o}C$ to $80^{o}C$), dipping in KI solutions, and many months operation in a vacuum. These test have clearly demonstrated that this material is chemically compatible with HgI_2 and an excellent barrier to HgI_2 diffusion and external corrosive materials.

Initial Parylene-C tests were performed on units whose coatings were applied by vendors, including the Union Carbide Service Center, in San Diego, CA. These initial tests were sufficiently promising to warrant building a small scale coating system for our laboratory that would allow optimization of the coating process and properties for this application. After some initial fine tuning, the system consistently produces water-clear coatings that appear superior to coatings

which are routinely obtained from commercial vendors. A practical thermal/vacuum process for HgI_2 crystal coating was developed to minimize exposure of the HgI_2 crystals to the vacuum before initiation of the polymer deposition. Early coating tests showed that exposure of the HgI_2 to the vacuum longer than 20 to 25 minutes prior to polymer deposition could lead to degraded final detector performance. A simple laser interferometer system has been developed to help control the thickness of the deposited layers.

As a check on reliability, in addition to visually examining the quality of test coatings under the microscope, tests for pin holes and permeability have been performed using a KI etching solution (potassium iodide solutions are extremely corrosive to HgI_2). A number of detectors with Parylene-C coatings were immersed in a 10% KI solution kept at 75-80°C. The thicknesses of the Parylene coatings ranged from 2 to 4 μm. After about one month, the solution was checked and determined to be over 40% KI, due to evaporation. The crystals were protected by the coating and no effects were found within this period of more than one month. An uncoated crystal would completely dissolve in a few minutes.

Recently we have started experimentation with other Parylenes because of the x-ray attenuation of Parylene-C. The chlorine content of the 2 micron thickness of Parylene-C produces an undesirable 10% attenuation of x-rays at 2.8 keV as well as significantly increasing attenuation at energies below 2 keV. Parylene-N (polymerized di-para-xylylene) contains no chlorine and so produces less x-ray attenuation for a given thickness. Because Parylene-N has significantly different evaporation/polymerization properties than -C, a new deposition process is being developed to produce the optimum protective coatings. Tests similar to that used for evaluating Parylene-C will be used with the new coatings.

Longevity Test

The long-term stability of the detectors is an important criterion in all applications, especially space missions. During the CRAF mission, the SEMPA instrument will be operating to perform x-ray analyses only a small fraction of the time. During flight from earth to the comet (several years) the instrument will be idle. However, during the entire mission, the instrument will be under vacuum conditions and exposed to thermal cycles induced by sun exposure and from other nearby science instruments being turned off and on. These storage and cycling conditions led us to a detailed evaluation of the durability of HgI_2 detectors.

In order to detect any inherent failure mechanisms in the detectors, we have remeasured the characteristics of HgI_2 x-ray detectors that had been fabricated as long as seven years previously in the very early stages of HgI_2 development at the University of Southern California (USC). These HgI_2 detectors had not been stored under any controlled conditions, but were simply kept in plastic boxes in the laboratory. We found that the energy resolutions of all detectors tended to improve slightly with age and storage time. The results seem to be evidence that there is no internal degradation mechanism at work in the HgI_2 crystal itself, over a time period of seven years. These detectors had all been protected by acrylic and/or silicone coatings. The apparent improvements in performance are probably due to improvements in the electronics used for the measurements.

For comprehensive testing of detectors and encapsulants under vacuum and/or thermal cycling conditions, a special apparatus with four separate detector chambers was constructed. Each chamber houses a detector and the preamplifier input field effect transistor, each attached to a separate thermoelectric coolers. All of the test chambers are connected to a common turbomolecular and ion pump manifold to achieve clean vacuums to 10^{-7} Torr, or equivalent to conditions in the SEMPA instrument.

For the past two years, four detectors encapsulated with Parylene-C, applied by a commercial source, have been undergoing testing in this system. The coating thicknesses are estimated to be about 4 μm. These detectors have also undergone bias cycling due to power failures at a rate of about once per month. Figure 4 gives a summary of energy resolution tests conducted at the Institute of Physics, USC. The tests on detectors N6-9F7 (in chamber #1) and N3-1F2 (in chamber #2) started in December 1986. The tests on detectors N6-9F8 (in chamber #3) and N3-1F1 (in chamber #4) started in November 1986. We have been monitoring the detectors' stability performance by measuring their energy resolution for the Mn k_α line, the electronic noise (pulser width) and their peak-to-background ratios. Curves 1 to 4 in Figure 4 show the energy resolution versus time for detectors N6-9F7, N3-1F2, N6-9F8 and N3-1F1, respectively. There have been no noticeable changes in the parameters of tested detectors. Certain variations in the results, which are greater than expected from statistics, are attributable to changes in the ambient temperature and the lack of stabilization of this parameter in our present system. During a one week short-term test, resolution was observed to vary about 25 eV. This variation could only be caused by system calibration errors, ambient temperature changes, and changes of the amplifier and cooling power supplies between measurements. From Figure 4 we can see that the long-term resolution variations of four detectors were not far beyond these short-term resolution changes, and that there is no clear systematic trend. Tests on these four detectors are still continuing.

Detector N3-1F2 (in chamber #2) and detector N6-9F8 (in chamber #3) were also subject to temperature cycling during part of the two-year test period. Two chambers were provided with external heaters in order to keep the temperature of the chamber body at about 40°C. Detectors were cooled with Peltier elements to about -20°C. A simple on/off programmable timer periodically switched power to the Peltier coolers, causing very rapid detector temperature changes between +40 and -20°C. The intervals of power on and off were from 1 to 2 hours. The actual detector temperatures changed between extremes in 1 to 2 minutes. These temperature extremes were chosen to approximate the limits which will be experienced by the SEMPA base plate during the CRAF mission. The results of this experiment, which was carried to 300 cycles, are presented in Table I. As can be seen from the table, there were no significant changes in the detectors' performance. Three hundred such temperature cycles exceeds the anticipated number of those cycles for the SEMPA instrument during the CRAF mission, and the temperature changes in the mission are expected to be more gradual and, therefore, less stressful for the detector.

The SEMPA x-ray probe system also serves to test the durability and longevity of HgI_2 detectors used in its routine operations. Routine instrument operations subject the detector to temperature, pressure and bias cycling tests, some of which are unintentional. After over one year of testing, the performance of a HgI_2 spectrometer is unchanged, showing energy resolution of about 200 eV (FWHM) for the Mn Kα line. The

detector is still undergoing continuous testing in the SEMPA instrument at the Jet Propulsion Laboratory (JPL).

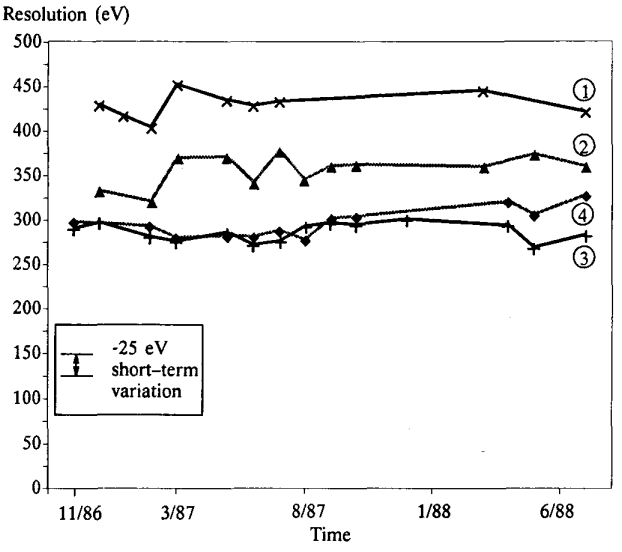

Figure 4. The long-term energy resolution variation for four tested HgI_2 detectors. The energy resolution (FWHM) was measured for the Mn k_α line. Curves 1 to 4 correspond to detectors in chambers #1 to #4, respectively.

TABLE I

DETECTOR TEMPERATURE CYCLING RESULTS

DATE	RESOLUTION/ PULSER/RATIO*	# OF CYCLES
DETECTOR N3-1F2		
08/03/87	348/296/151	0 CYS
08/05/87	382/295/153	12 CYS
08/13/87	367/290/154	60 CYS
08/26/87	354/286/151	**150 CYS
09/21/87	363/291/131	300 CYS
DETECTOR N6-9F8		
08/03/87	298/259/294	0 CYS
08/07/87	298/258/238	2 CYS
08/13/87	295/248/261	5 CYS
08/17/87	301/248/243	10 CYS
08/24/87	298/243/356	18 CYS
09/01/87	299/249/253	**72 CYS
09/14/87	292/243/245	150 CYS
12/02/87	303/245/245	300 CYS

* RESOLUTION OF Mn-k_α LINE (FWHM) IN (eV)/ PULSER WIDTH (FWHM) IN (eV)/ PEAK TO BACKGROUND RATIO.

** FULL CYCLE WAS CHANGED FROM 4 HOURS TO 2 HOURS.

Radiation Damage of HgI_2 Detectors

Radiation damage to semiconductor detectors is known to occur during space flight. Over a multi-year mission, the accumulated damage from cosmic rays and their products can result in changes in the operating properties of solid state detectors. In severe cases the detector can be damaged to a point that it is no longer useful.

There is little data in the literature on radiation damage of HgI_2 x-ray detectors. Some very preliminary results were obtained with gamma detectors by Recchetti et. al.(9). Recently, we have performed some initial, controlled tests of mercuric iodide x-ray detectors to assess their vulnerability to proton radiation.

For that purpose six medium quality HgI_2 x-ray detectors protected by PMMA coatings were selected and their characteristics (leakage current, FWHM of Fe-55 x-ray line, electronic noise, peak to valley ratio of x-ray line, etc.) were measured before the irradiation. All tests were done using the same resistor feedback preamplification system. Optimum electronic noise level and energy resolution were not primary considerations in these initial tests. The detectors were exposed to an external beam of 10.7 MeV protons from the Argonne National Laboratory accelerator, to fluences up to 10^{12} protons/cm^2, to see at what point changes in detector performance could be observed. The fluences were accumulated during periods lasting several minutes and represent the worst case for potential detector damage. Usually during transit in space, the rates of irradiation from cosmic rays are much lower, and the detector may be self-annealing during that period. Within one to two weeks after the irradiations, the same characteristics were measured again. Table II lists parameters measured before and after each detector's irradiation. From these results, it is clear that all six detectors survived the irradiation without any appreciable change in their performance. The small observed changes can be explained in terms of variations in the test conditions.

TABLE II

LIST OF DETECTOR PARAMETERS BEFORE AND AFTER PROTON IRRADIATION

Det.#	Dose*	Resolution* Before	Resolution* After	Pulser* Before	Pulser* After	PK/BKG* Before	PK/BKG* After	Leakage* Before	Leakage* After
N8-8F3	6.6×10^9	394	409	311	370	46:1	43:1	0.3	1.3
E11-5LF3	6.6×10^9	472	496	366	369	66:1	67:1	4.0	2.3
N13-6F1	4.8×10^{10}	490	426	422	350	193:1	248:1	3.7	4.8
S8-2SF5	4.8×10^{10}	483	491	436	409	215:1	196:1	0.2	0.1
N8-8F2	10^{12}	473	459	437	380	152:1	220:1	0.8	0.8
K7-11DF2	10^{12}	532	502	385	394	165:1	152:1	4.0	3.9

* Dose: protons/cm^2
Resolution: Mn-k_α line (FWHM) in (eV)
Pulser: pulser with (FWHM) in (eV)
PK/BKG: peak to background ratio
Leakage: leakage current (PA).

In some space applications, the expected accumulated doses could be even higher than the above doses. It will be important, therefore, to extend the fluences and energies to which the HgI_2 x-ray detectors are tested for radiation damage susceptibility. Because ten MeV protons represents the low energy range of cosmic rays, it would be extremely interesting to study the effects of irradiating detectors with higher energy protons as well as with other types of radiation that may have different effects.

Conclusions

All of the experimental and test results described in this paper have shown that HgI_2 detectors are suitable for the ordinary requirements of energy dispersive detectors in x-ray spectroscopy systems. The HgI_2 detectors have shown excellent durability during two-year longevity tests under difficult conditions. Detectors have also shown impressive resistance to proton irradiation damage. Energy resolution measurements better than 200 eV in a practical, noncryogenic detector system represent the achievement of a major milestone, and help confirm the feasibility of utilizing HgI_2 detectors for applications requiring good energy resolution at x-ray energies from 1 to 10 keV.

The research work on improvements in HgI_2 detector fabrication and amplification electronics must continue. We expect to achieve improved low energy detector response by using thinner coatings of protective encapsulants of lower atomic numbers. The long-term life testing will also be repeated with the new coatings. Bias cycling and extended range temperature cycling are planned. Longer exposures and higher proton energies for radiation damage testing are planned in the next phase of this project.

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References

[1] J. S. Iwanczyk, A. J. Dabrowski, and G. C. Huth, "A Study of Low-Noise Preamplifier Systems for use with Room Temperature Mercuric Iodide (HgI_2) X-Ray Detectors," IEEE Transactions on Nuclear Science, Vol. NS-28, No. 1, pp.579-582, February 1981.

[2] J. S. Iwanczyk, "Advances in Mercuric Iodide X-Ray Detectors and Low Noise Preamplification Systems," Presented to the Sixth International Workshop on Compound Semiconductors for Room Temperature X-Ray and Nuclear Detectors, Davos, Switzerland, 14-19 September, 1987. Text will appear in the Conference Proceedings issue of Nuclear Instruments and Methods (1988).

[3] J. G. Bradley, J. M. Conley, A. L. Albee, J. S. Iwanczyk, A. J. Dabrowski, and W. K. Warburton, "Practical Application of HgI_2 Detectors to a Space-Flight Scanning Electron Microscope," Presented to the Sixth International Workshop on Compound Semiconductors for Room Temperature X-Ray and Nuclear Detectors, Davos, Switzerland, 14-19 September, 1987. Text will appear in the Conference Proceedings issue of Nuclear Instruments and Methods (1988).

[4] J. M. Conley, J. G. Bradley, C. E. Giffin, A. D. Tomassian, and A. L. Albee, "Development of a Miniature Scanning Electron Microscope for in-flight analysis of comet dust," Microbeam Analysis, pp.177-181, 1983.

[5] R. K. Hart, A. L. Albee, A. A. Finnerty and R. Frazer, "A Mini-scanning Electron Microscope and Particle Analyzer for Space Applications," Scanning Electron Microscopy, I, pp. 96-104, 1981.

[6] Jet Propulsion Laboratory, "The Comet Rendezvous Asteroid Flyby Mission: A Search for Our Beginnings, National Aeronautics and Space Administration," Jet Propulsion Laboratory Publication JPL 400-320, 1987.

[7] W. K. Warburton and J. S. Iwanczyk, "Advanced Mercuric Iodide Detectors for X-Ray Microanalysis," Scanning Microscopy Supplement 1, pp. 135-150, 1987.

[8] J. S. Iwanczyk, A. J. Dabrowski, G. C. Huth, J. G. Bradley, J. M. Conley, A. L. Albee, "Low Energy X-Ray Spectra Measured With a Mercuric Iodide Energy Dispersive Spectrometer in a Scanning Electron Microscope," IEEE Transactions on Nuclear Science, Vol. NS-33, No. 1, pp. 355-358, February, 1986.

[9] F. S. Becchetti, R S. Raymond, R. A. Ristinen, W. F. Schnepple and C. Ortale, "Mercuric Iodide (HgI_2) Semiconductor Devices as Charged Particle Detectors," Nucl. Instr. and Meth. 213 (1983) 127-132.